Glass and Ceramics *Vol. 55, Nos 3 – 4, 1998*

UDC 666.792.6:546.621

PRODUCTION OF NITRIDE CONTAINING MIXTURE IN OXIDATION OF ALUMINUM POWDER IN AIR

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Translated from Steklo i Keramika, No. 3, pp. 24 – 25, March, 1998.

The authors investigate the composition of the combustion products of mixtures of freely poured aluminum powders: ADS-1 industrial powder and ultradisperse aluminum powder produced by electric detonation of conductors in argon. It is demonstrated that the end products containing ~50% AlN are easily destroyed sinters and can be used as the main component of the mixture for production of the nitride-containing ceramics.

One of the problems in nitride (AlN, BN, Si_2N_4) sintering is the search for sintering additives (of the Y_2O_3 type) which provide a liquid phase without deteriorating the properties of the ceramics [1]. At the same time, these additives take part in the formation of transitional layers between the nitride particles, thus improving the mechanical properties and increasing the density of the product.

It was earlier demonstrated that under certain conditions of combustion of ultradisperse powder (UDP) of aluminum in air, it is possible to obtain powders with a mass content of aluminum nitride above 50% [2]. An analysis of the processes taking place during combustion with free access of air suggests that the emerging aluminum nitride is protected on the surface by oxide phases: γ- and α- oxides of aluminum. Otherwise, in the course of the final combustion (oxidation) the nitride would be transformed into oxide: the temperature drop from the peak (1120 – 1400°C) to 600 – 800°C lasts tens of seconds, which is sufficient for oxidation of the nitride. According to thermodynamics, the AlN oxidation process is exothermic: the enthalpy of its formation is ΔH_f° (298 K) ≈ 335 kJ/mole. A serious obstacle for the practical application of formation of nitride in combustion in air is the short supply of ultradisperse powders; therefore, we investigated the effect of dilution of UDP with ASD-1 industrial aluminum powder.

The present paper describes the results of the investigation of the formation processes and compositions of nitride-containing powders resulting from combustion of aluminum powder in air. The mixtures of aluminum UDP and ASD-1 powder were prepared by dry mixing. The aluminum UDP was produced by electric detonation of conductors in argon [3]. The UDP particles had a spherical shape, and the distribution of particles by size was close to the normal logarithmic value. The content of aluminum metal was 91.2%, the average surface diameter of the particle approached 0.08 μm. The average surface diameter of ASD-1 powder particles was 80 μm, and the metallic aluminum content was 99.5%. The apparent density of the aluminum ADP was 0.2 g/cm³, and that of ASD-1 powder was 1.6 g/cm³.

The mixtures of aluminum UDP and ADS-1 powder were placed on a metal base (steel) and a conical shape was imparted to the charge. The combustion process was initiated with a nichrome spiral through which a pulse of current was transmitted. The sample temperature in the course of combustion was controlled by a chromel-alumel thermocouple (conductor diameter of 0.5 mm), and its hot junction, protected with a ceramic cap, was inserted in the sample. The need to use large-diameter conductors and to protect the

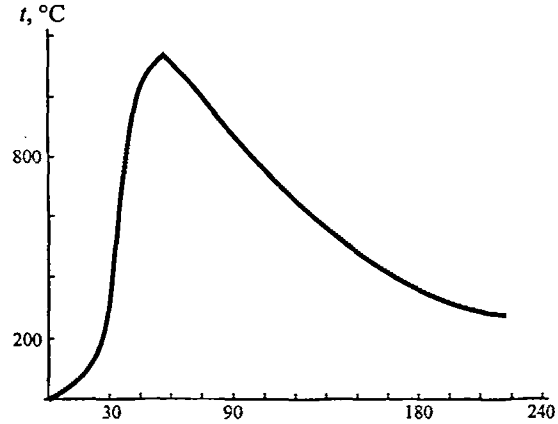

Fig. 1. Typical curve of the relative variation of the sample temperature in the course of combustion.

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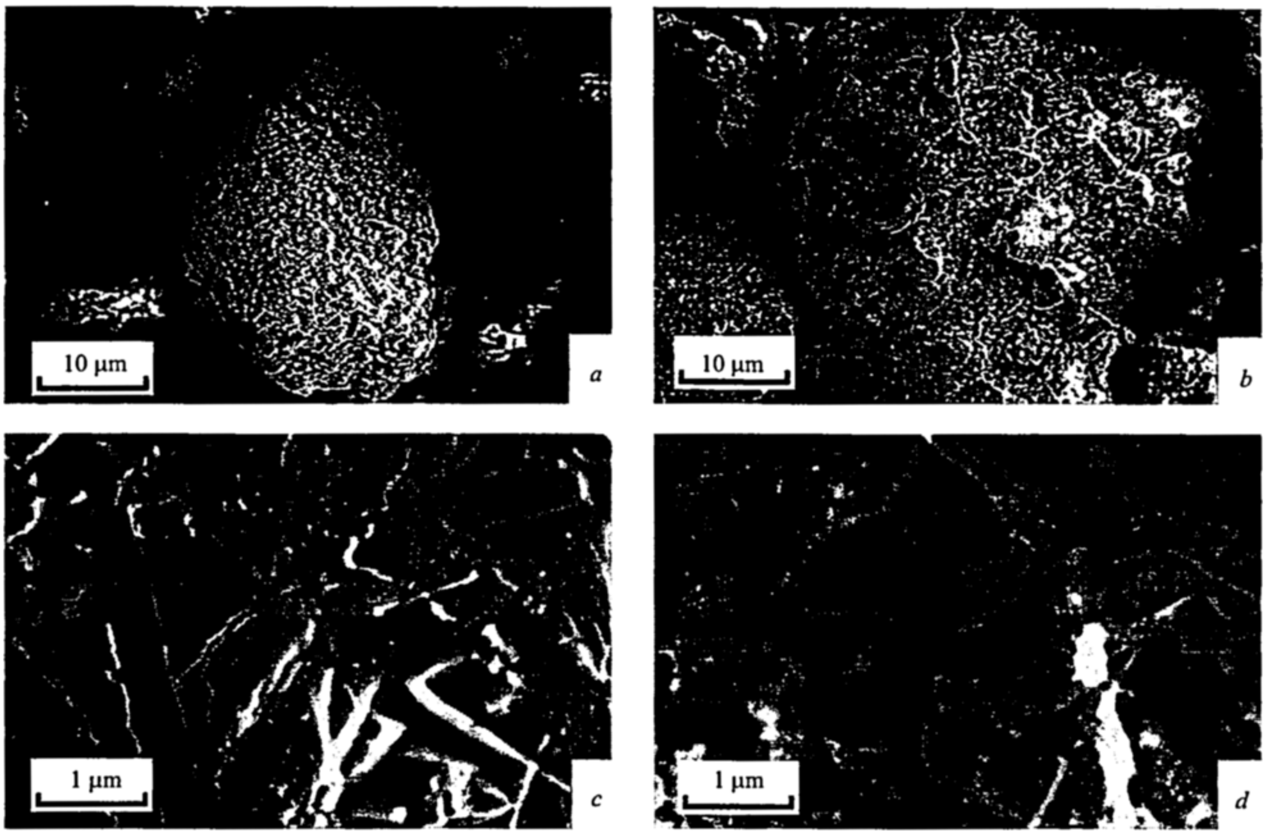

Fig. 2. Photos of the powders taken with the electron microscope. Initial mixture composition: *a, c*) 100% aluminum UDP; *b, d*) 40% aluminum UDP, 60% ASD-1 powder.

junction with a ceramic cap is due to the high activity of aluminum vapor. At the same time, the large diameter and the junction protection produce a time lag and underestimation of the absolute values of the temperature measurements. Therefore, we used these data for a comparative evaluation of the combustion of different mixtures. The temperature variation in the process of combustion was recorded with the KCP-4 recorder. The combustion products after cooling were crushed and underwent x-ray phase analysis (DRON-3.0 diffractometer, $Cu_{K\alpha}$-radiation).

Having been initiated, the combustion process proceeds as described in [2] in two stages: slow combustion at a relatively low temperature, and a rapid increase in the temperature accompanied by a bright glow. Fig. 1 shows a typical curve of the sample temperature in the course of its combustion in air. The powder mixtures containing up to 50% (inclusive) ASD-1 powder burned up in two stages with production of AlN. With a further decrease in the UDP content in the mixture, the combustion process proceeded in a single stage, or could not be initiated due to the increased apparent density and heat removal. An increase in the UDP dispersity makes it possible to further reduce its content in the mixture

According to the data of the x-ray phase analysis, AlN was observed in the resulting products in all samples whose combustion was accompanied by a sharp increase in the temperature (the second stage). AlN as an independent phase has 100% reflections on the x-ray diagram. In the case of formation of AlN, the end product contains 20 – 40% α-Al_2O_3 which is caused by the high combustion temperature. The only exception is the sample containing 40% aluminum UDP and 60% ASD-1 powder for which the 100% reflection is that of metallic aluminum ($d = 2.338$ Å). Apparently, an AlON phase is present in the combustion products containing AlN. It is supported by the higher intensity of the aluminum nitride reflection ($d = 2.37$ Å) close to the reflection of oxynitride ($d = 2.39$ Å). The difficulty in identifying the AlON phase is due to the coincidence of the main reflections of AlON and γ-Al_2O_3.

The study of the combustion product employing the scanning electron microscope (JEOL-740 microscope) showed (Fig. 2) it to consist of spheroidal sinters 20 to 200 μm in diameter joined in easily broken associates of millimeter size (Fig. 2*a, b*). At a greater magnification it can be seen that part of the combustion products consists of elongated and needle-shaped formations, where their length ranges from several millimeters to tens of millimeters, and their thickness is less than 1 μm (Fig. 2*c*). Among the elongated formations, needle-like shapes of thickness below 0.2 – 0.1 μm are occasionally found, although the initial

powder particles were spherical. Apparently, this significant modification of the particle shape is associated with mass-transfer processes during combustion.

Thus, in the course of combustion of the mixture of aluminum UDP and ASD-1 powder, the composite powders containing AlN (~ 50%), γ-Al_2O_3 and residual aluminum (~ 5 – 7%) were obtained. After some preparation, such powders can be used as a mixture for the production of nitride-containing ceramics.

REFERENCES:

1. G. V. Samsonov, O. P. Kulik, and V. S. Polishchuk, *Production and Methods of Analysis of Nitrides* [in Russian], Naukova Dumka, Kiev (1978).
2. A. P. Il'in and L. T. Proskurovskaya, "Two-stage combustion of aluminum ultradispersed powder in air," *Fiz. Goreniya Vzryva*, No. 1, 71 – 72 (1990).
3. A. P. Lyashko, G. G. Saveliev, and D. V. Tikhonov, "Morphology, phase composition, and oxidation of powders obtained by electric detonation of brass wires," *Fiz. Khim. Obrab. Mat.*, No. 6, 127 – 130 (1992).